Thermal conductivity of CNT water based nanofluids: Experimental trends and models overview

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Abstract: We report in this communication thermal conductivity measurement of carbon nanotubes water-based nanofluids. We consider in particular the influence of nanoparticle volume fraction, temperature, carbon nanotube aspect ratio and different kind of surfactant (SDBS, Lignin, Sodium Polycarboxylate) on thermal conductivity enhancement of nanofluids. In addition, various theoretical thermal conductivity models are presented in an attempt to correlate the experimental data.

Keywords: CNT nanofluids, Thermal conductivity, Experiments, Models, Temperature, Nanoparticle aspect ratio, surfactant

1. Introduction

Since many years, it has been demonstrated that nanofluids (NF) are promising candidates as heat transfer fluids in heat exchangers, cooling devices, and solar collectors [1-3] due to their enhanced thermal properties. Nanofluids are obtained by dispersing a small amount of nanoparticles with high intrinsic conductivity within currently used base fluids, such as water, ethylene glycol, oil, … Among the numerous studied systems, carbon nanotubes (CNT) appear very interesting due to their high thermal conductivity in comparison to Al_2O_3, CuO and TiO_2. In addition, as nanofluid viscosity is related to resistance to flow and pumping power in energy systems, it is better to use low viscosity base fluid. Thermal conductivity is an important physical properties which can be used to evaluate the efficiency of nanofluid as coolants [4,5], predict heat transfer within exchangers [6,7] and natural convection in cavities as well [8].

Hence, the purpose of the present communication is to report the thermal conductivity (TC) measurement of water-based nanofluids containing carbon nanotubes. CNT consist here in MWCNT stabilized by different surfactants. Actually, in addition to mechanical stirring and sonication, due to the hydrophobic behavior of CNT their dispersion and stability within water is obtained through the use of surfactant. We have considered here the influence of SDBS (sodium dodecylbenzenesulfonate), lignin, which is a by-product of paper industry and sodium polycarboxylate respectively as stabilizers. The effects of nanoparticle volume fraction within the range 0.55-0.0055%, temperature varying between 20 and 40°C, and nanoparticles aspect ratio (90; 160) on the thermal conductivity enhancement is first presented and discussed.

Moreover, various theoretical approaches and empirical models are presented considering the influence of nanofluid composition and impact of several mechanisms such as volume fraction, temperature, nanoparticle shape and aspect ratio, Brownian motion, carbon nanotubes curving and wrapping, interfacial thermal resistance of CNT, … Finally, experimental results are discussed and compared to theoretical predictions.

2. Materials and experiments

2.1 Materials

Four types of nanofluids were here selected and investigated. They have been partially studied in our previous works [5,9-12]. Nanofluids consists of MWCNT (purity of 90%) dispersed a mixture of deionized water and surfactant. Table 1 summarizes the composition and the properties of the different nanotubes and nanofluids. In all cases, an initial starting suspension with 1% in weigth fraction of nanotubes and 2% in weight fraction of surfactant was prepared by Nanocyl. Nanofluids with lower volume fraction were obtained from serial dilution of the starting suspension, as reported earlier [9-12], conserving constant surfactant/carbon nanotubes weight ratio of 2. Finally, the whole volume fraction range investigated varies between 0.0055% and 0.55%. As evidenced by table 1, impact of average CNT aspect ratio and surfactant nature are presently studied.

Table 1. Nanotubes and nanofluids properties

	N_1	N_2	N_3	N_4
Nanotube average diameter d (nm)	9.2	9.2	9.2	11.4
Nanotube average length l (µm)	1.5	1.5	1.5	≈1
Average aspect ratio (r=l/d)	160	160	160	90
Density (kg/m^3)	1800	1800	1800	2050
Carbon purity (wt.%)	90	90	90	90
Sphericity / n (H-C model)	0.24 / 12.5	0.24 / 12.5	0.24 / 12.5	0.29 / 10.23
Surfactant	SDBS	Lignin	Sodium polycarboxylate	Sodium polycarboxylate

2.2 Thermal conductivity measurement

A KD2 Pro thermal property analyzer (Decagon Devices Inc.), which is based on the transient hot wire method, was used to evaluate the thermal conductivity of both nanofluids and base fluids (deionized water and surfactant). The experimental set-up for thermal conductivity measurement was previously used in [11,12]. Its accuracy was also previously evaluated with distilled water, leading to a maximum standard deviation of 3.5%. During the measurement, both the sample and the probe were first maintained 30 min at the required temperature. Then, ten measurements were recorded with 5 mins of interval. In the following, the thermal conductivity values also consist of an average of the ten measures performed at 20, 30 and 40°C respectively for each volume fraction.

3. Thermal conductivity models for CNT nanofluids

Several theoretical correlations have been developed in the past to predict the thermal conductivity enhancement of CNT nanofluids. The relevance of many correlations have been presented and discussed in recent works [13,14]. Here, we intend to compare some of these correlations with our experimental data, considering different factors that can contribute to the theoretical frame of thermal conductivity enhancement.

At first sight, the thermal conductivity of nanofluids can be estimated by Hamilton and Crosser model [15]. This model, defined by equation (1), can be used when the thermal conductivity of the particles is at least 100 times higher than the one of the liquid phase.

$$k_{nf} = \frac{k_p + (n-1)k_{bf} + (n-1)(k_p - k_{bf})\phi}{k_p + (n-1)k_{bf} - (k_p - k_{bf})\phi} k_{bf} \qquad (1)$$

In the previous equation, k_{nf}, k_p and k_{bf} are the thermal conductivity of nanofluids, nanoparticles and base fluid respectively in W/mK, ϕ is the volume fraction and n is a shape factor linked to nanoparticles sphericity such as $n=3/\psi$. Due to rod shape of nanotubes contained within the nanofluids, the sphericity ψ was here calculated from the average length and diameter of the nanotubes investigated. This leads to n values reported in Table 1 for each tested nanofluids. Table 1 shows that n values vary following CNT aspect ratio and differs from 6, as generally considered with cylindrical particles. It is noted that equation (1) does not support directly the influence of temperature.

A thermal conductivity model for nanofluids containing nanotubes taking into account the effect of diameter and aspect ratio of nanotubes as well Brownian effect due to temperature has been developed by Walvekar et al. [16]. This model derives from a model initially introduced by [17] and writes as follows.

$$k_{nf} = k_{bf}\left[1+\frac{k_p\left(\dfrac{2\phi(r_p+l_p)}{r_p l_p}\right)}{k_{bf}\left(\dfrac{3(1-\phi)}{r_{bf}}\right)}\right]+\frac{C\phi(T-T_0)}{r_p{}^2 l_p{}^2\mu_{bf}}\ln\left(\frac{l_p}{d_p}\right) \qquad (2)$$

In this equation, r_p is the radius of nanoparticles (m), l_p is the length of the nanoparticles (m), r_{bf} is the radius of base fluid molecule (m), μ_{bf} is the base fluid viscosity (Pa.s), and T is the temperature (K). It is worth noting that viscosity of the different base fluids was previously measured [9,12] or additional measurements were performed following the procedure described in [9]. As reported earlier [12,16], the first part of the model is mainly related to volume fraction, shape and aspect ratio of nanoparticles and thermal conductivities of both nanoparticle and base fluid. The second term is related to Brownian movement of nanoparticles linked to temperature and base fluid viscosity. T_0 is taken as 273°K which corresponds to the reference temperature below which Brownian motion becomes negligible. C depends on Boltzmann constant as $C=85k_B^2/72\pi^2$ [16].

The effects of carbon nanotube curving and wrapping was investigated in [17]. Such phenomena depend on base fluids, nanotubes dimensions and presence of surfactants. In this work, several equations were developed to predict TC enhancement of CNT-based nanofluids from a distribution based modelling technology and the use of probability density functions (denoted p(x)). The authors showed that among the models developed, the uniform distribution model, denoted UDM (equation 3, obtained with p(x)=3) and the linear increase distribution model, denoted LIDM, (equation 4 with p(x)=18(1/3-x)) are able to capture the TC enhancement of CNT-based nanofluids. So, the effect of moderate curving and wrapping is captured by the following equation under the consideration of $k_{bf}/k_p \ll 1$.

$$k_{nf} \approx 1+\phi\ln\left(\frac{27k_p}{16k_{bf}}\right)k_{bf} \qquad (3)$$

For less curving and wrapping nanotubes, TC of nanofluid can be obtained from ($k_{bf}/k_p \ll 1$)

$$k_{nf} \approx 1+6\phi\left[1+\frac{1}{3}\ln\left(\frac{256k_p}{19683k_{bf}}\right)\right]k_{bf} \qquad (4)$$

Finally, Nan et al. [19] proposed a TC model for carbon-based nanofluids taking into account the effect of interfacial resistance based on multiple scattering theory and the EMT model of Maxwell, considering also the Kapitza resistance at the CNTs medium. This model was expressed as

$$k_{nf} = \frac{3+\phi(\beta_{11}+\beta_{33})}{3-\phi\beta_{11}}k_{bf} \qquad (5)$$

where $\beta_{11} = \dfrac{2\left(k^c{}_{11}-k_{bf}\right)}{k^c{}_{11}+k_{bf}}$ and $\beta_{33} = \dfrac{k^c{}_{33}}{k_{bf}}-1$

In these equations, $k^c{}_{11}$ and $k^c{}_{33}$ denote the equivalent TC along transverse and longitudinal axes respectively of a thin interfacial thermal layer and depend on CNT dimensions and Kapitza radius a_k [13,19].

It is shown that all these equations require properly determination of both TC and dimensions of nanotubes, and the properties of base fluid. It should also be mentioned that the possible effect of particle agglomeration do not appear within the equations. As reported earlier, the TC of CNT is 3000 W/mK.

4. Results and discussion

As indicated in section 2.1, due to constant surfactant/carbon nanotubes weight ratio, the quantity of surfactant increase with CNT volume fraction. Moreover, three kinds of surfactant were used here. So, the effect of surfactant content and nature is first investigated reporting the TC of base fluid in function of surfactant quantity. This is shown by Figure 1 at 30°C. As reported earlier [12], it is observed that TC of base fluid decreases when the amount of surfactant is increased, and that the TC is lower than the one of deionized water at this temperature. This means that surfactants penalize the TC of NF. It is also shown that this effect is independent of surfactant type, within the experimental uncertainty. Similar tendencies were obtained at lower and higher temperatures (20 and 40°C), TC being decreased and increased when the temperature is decreased and increased respectively.

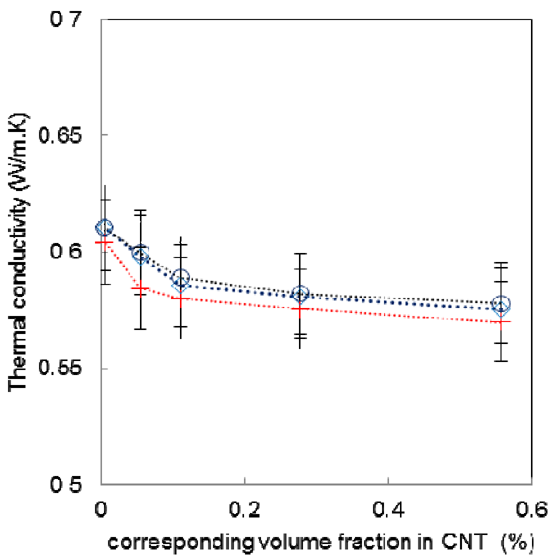

Figure 1. Thermal conductivity of base fluids at 30°C – Impact of nature and quantity of surfactant (open circle: sodium polycarboxylate ; open diamond-shaped: lignin ; cross: SDBS)

Figure 2 shows the effect of temperature and volume fraction on the thermal conductivity enhancement of NF for two nanofluids N_3 and N_4. A similar behaviour was also noticed for N_1 and N_2. As often reported in literature, TC of NF increases when both the CNT nanotube volume fraction increases and temperature as well. The TC quickly enhances for lower CNT volume fraction, in particular at 30 and 40°C. In addition, at these temperatures, TC do not follow a linear trend with volume fraction.

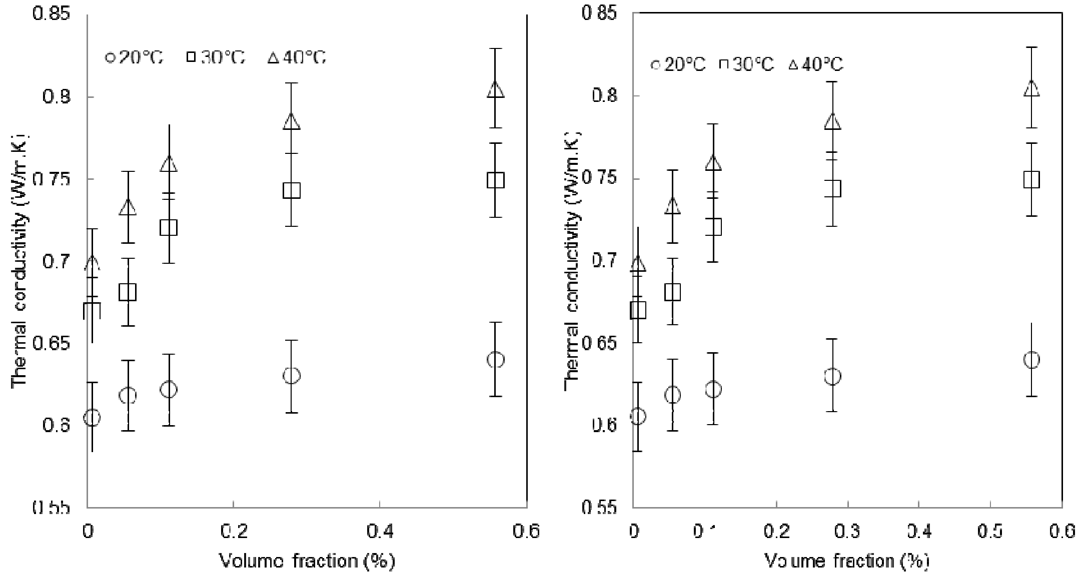

Figure 2. Thermal conductivity of N3 (left) and N4 (right) – Impact of CNT volume fraction and temperature

The thermal conductivity of NF is reported in Figure 3(left) for N_1, N_2 and N_3 at 20°C evidencing the effect of surfactant on TC. While TC of base fluid decreases with the amount of surfactant, figure 3(left) shows that TC of nanofluids increases with CNT volume fraction. When temperature is increased, similar trend is also noticed. The figure also shows that surfactant nature do not influence TC enhancement except at higher volume fraction for SDBS. The effect of CNT aspect ratio is shown in Figure 3(right) at 30°C (at 20°C this effect is negligible; the effect is increased at 40°C). Higher the aspect ratio, higher is the TC enhancement at high volume fraction. So, this agrees well with previous published results. However, the maximum deviation, which is obtained at 0.55% and 40°C, is only around 5%.

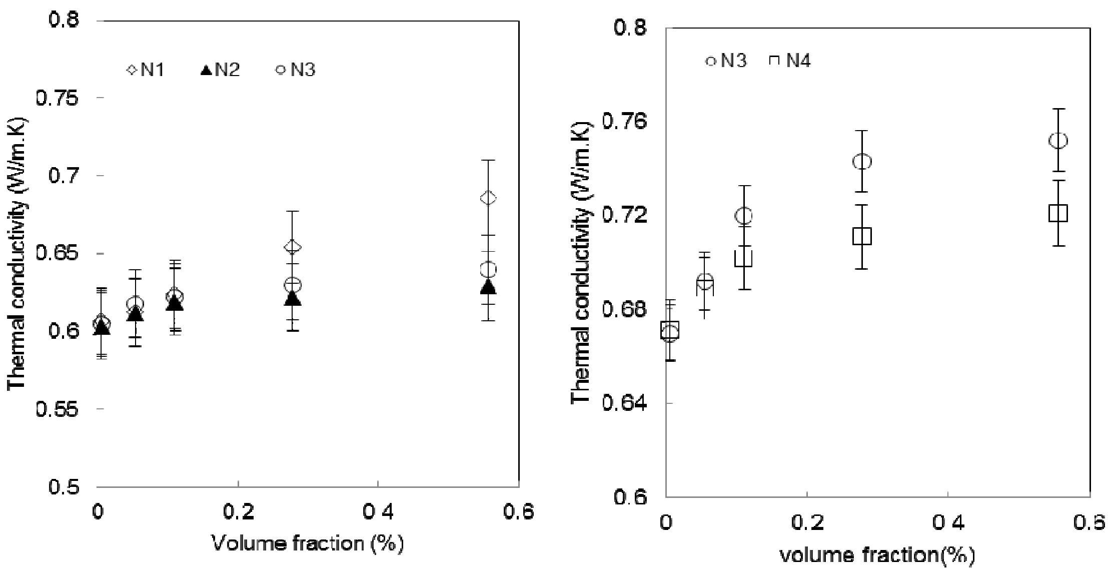

Figure 3. Effect of surfactant (left) and CNT aspect ratio (right) on thermal conductivity of nanofluids

Based on these experimental findings, we focus now on the comparison between experimental data and theoretical correlations presented above considering mainly the influence of volume fraction, temperature and CNT aspect ratio. So, N_3 and N_4 nanofluids are only discussed in the following.

As for TC, Figures 4 and 5 show that relative TC (RTC) of nanofluids increases quite linearly at 20°C. When the temperature is increased, RTC of nanofluids sharply increases at low volume fraction up to 0.111%. Then, the increasing goes up more slowly. This also evidences that the penalizing effect of surfactant on TC of water reported before is not predominant in comparison with TC of nanotubes. As expected also, the better RTC is achieved for both the higher volume fraction and temperature.

It is also observed from both figures that the theoretical predictions are much lower than the experimental data except for Walvekar model. However, some of the proposed models are able to correlate the experimental data at 20°C (within the experimental uncertainty), the best correlation being obtained with the LIDM model. When the temperature is increased, the models greatly deviate in comparison with experimental data and do not report in particular the great enhancement in TC at low volume fraction in nanotubes.

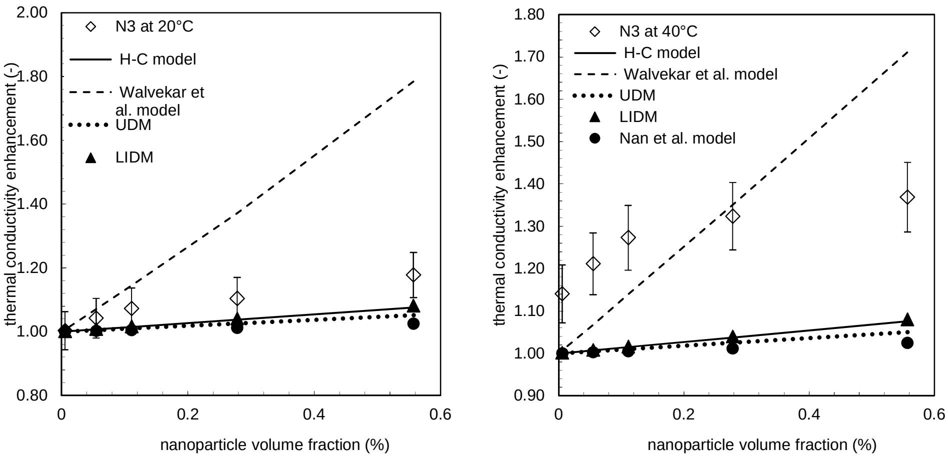

Figure 4. Relative TC enhancement of N_3 in function of nanoparticle volume fraction at 20°C (left) and 40°C (right) – Comparison with theoretical correlations

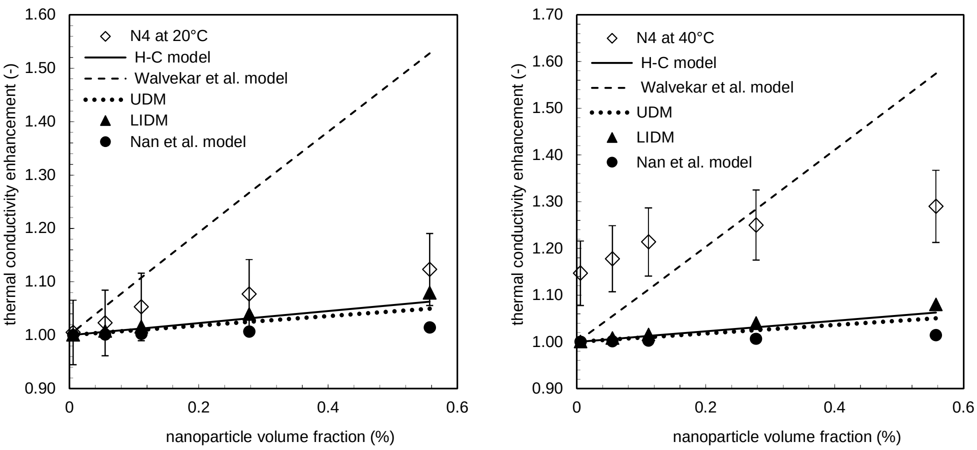

Figure 5. Relative TC enhancement of N_4 in function of nanoparticle volume fraction at 20°C (left) and 40°C (right) – Comparison with theoretical correlations

Finally, this experimental study evidences the potential of carbon nanotubes water-based nanofluids as heat transfer media and coolants for thermal applications due to the great enhancement of TC even at low volume fraction. In addition, none of the selected TC models properly predict TC enhancement of carbon nanotubes water-based nanofluids within the entire range of volume fraction, temperature and nanotubes aspect ratio. While several factors affecting TC have been considered, there is little difference between the studied TC models apart at the higher volume fraction in nanotubes. However, the results suggest that curving and wrapping and size effect of CNT appear at high volume fraction and low temperature of 20°C. For higher temperatures, conclusions are not so far easy and one factor cannot be only used to predict RTC enhancement from theoretical point of view.

5. Conclusion

A parametric experimental study of thermal conductivity of water-based carbon nanotubes was presented. In particular, we have considered the influence of surfactant used to stabilize the nanotubes, carbon nanotubes volume fraction and aspect ratio and temperature as well. It was observed that TC enhancement of CNT nanofluids increase with volume fraction and temperature. The thermal conductivity enhancement is really

significant at very low volume fraction, in particular at 30 and 40°C. The effect of the used surfactants on TC enhancement of nanofluids is weak, TC is also weakly affected by the influence of CNT aspect ratio considered here.
The comparison between experiments and models show that theoretical predictions presented above cannot clearly capture the TC enhancement of CNT water based nanofluids presently investigated within the entire range of volume fraction considered and temperature. This evidences a need both to develop appropriate model for TC enhancement prediction of CNT nanofluids and measure TC of this kind of nanofluids before performing numerical studies in heat exchangers and cavities.

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